

Third minima in actinides - do they exist?

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We study the existence of third, hyperdeformed minima in a number of even-even Th, U and Pu nuclei using the Woods-Saxon microscopic-macroscopic model that very well reproduces first and second minima and fission barriers in actinides. Deep ($3 \div 4$ MeV) minima found previously by Ćwiok et al. are found spurious after sufficiently general shapes are included. Shallow third wells may exist in $^{230,232}\text{Th}$, with IIIrd barriers ≤ 200 and 330 keV (respectively). Thus, a problem of qualitative discrepancy between microscopic-macroscopic and selfconsistent predictions is resolved. Now, an understanding of experimental results on the apparent third minima in uranium becomes an issue.

The topic of third minima in actinides, supposedly more deformed than the superdeformed (SD) ones, and for this reason sometimes called "hyperdeformed", is surrounded by some uncertainty. Their existence is inferred from an analysis of the observed transmission resonances in the prompt fission probability in (n,f), (d,pf), (γ ,f) and (e,f) reactions. Using one-dimensional models for tunneling one tries to fit the data by considering various possibilities of rotational or vibrational states, either in the second or the third well, and choosing unknown barrier parameters. The best fit gives energy at the third barrier and minimum, and possibly the moment of inertia in the IIIrd well. Additional check of the hypothesis may be obtained by measuring fission fragment angular distribution. First interpretation of the third minima in thorium [2] was supported by later studies [3–7] in $^{230,231,232,233}\text{Th}$ which found them rather shallow ≈ 300 keV. Recent experiments of the Debrecen-Munich group report third minima in a number of nuclei ($^{232,234,236}\text{U}$, ^{232}Pa) [8–11]. Based on resonance energies inbetween the first and the (higher) second barrier and their small widths, they infer a third barrier at least as high as the second one and deep third minima. There are not any direct measurements of the third minima so some reservations concerning the interpretation would be in order.

Theoretical predictions of the third minima in actinides present somewhat confusing picture. While the presence of the second minima at deformation close to $\beta_2 = 0.6$ is a common feature of both macroscopic-microscopic (since Strutinsky [1]) and self-consistent calculations, the third minima appear or disappear from calculation to calculation. The first calculated IIIrd minima [12, 13] were predicted shallow, 0.5 to 1.5 MeV, with the third barrier higher than the second one in thorium. Bengtsson et al. [14] had very shallow minima (0.5 MeV or less), with the lower third barrier. Qualitatively different results, with prominent 3-4 MeV deep, and often multiple, third minima, also in U and heavier nuclei, were presented by S. Ćwiok et al. [15–17]. These calculations used a richer set of axially symmetric deformations than

the previous ones, up to the multipolarity $\lambda = 7$. Results of the Hartree-Fock calculations indicate that presence or absence of third minima depends very much on the effective interaction. However, minima in U and Pu, if they appear at all, are rather shallow and nearly degenerate with, or even lower than, the ground states [21]. Therefore, they do not fit Debrecen-Munich results. On the other hand, calculations with the Gogny force [22] and SkM* [23] (Hartree-Fock plus BCS) predict shallow minima in some Th isotopes, compatible with Th data [5], while the other work [24] provides shallow minima at the top of the first barrier, not of the type suggested by the experimental papers.

The aim of this work was to check the existence of the third minima in actinides within the Woods-Saxon microscopic-macroscopic model used in [15, 16] by extending the number of included deformations. An impulse to this came from first repeating old calculations and realizing that they produce IIIrd minima also in many heavier actinides, in which they were not found experimentally. Moreover, the calculated minima with larger octupole deformations have quadrupole moments (calculated assuming the sharp-surface distribution and the radius parameter 1.16 fm) $Q \sim 170$ b, disturbingly close to the scission region. The contemplated possibility was that some IIIrd minima in [15, 16] are just intermediate configurations on the scission path, whose energy was calculated erroneously because of limitations of the admitted class of shapes. *Method* Third minima were searched by exploring energy surfaces in a region of deformations beyond the second minima. The energy is calculated within the microscopic-macroscopic method with a deformed Woods-Saxon potential. Nuclear shapes are defined in terms of the nuclear surface [27]

$$R(\theta, \varphi) = c(\{\beta\})R_0(1 + \sum_{\lambda>1} \beta_\lambda Y_{\lambda 0}(\theta, \varphi) + \sum_{\lambda>1, \mu>0, \text{even}} \beta_{\lambda\mu} Y_{\lambda\mu c}(\theta, \varphi) + \sum_{\lambda>1, \mu>0, \text{odd}} \beta_{\lambda\mu} Y_{\lambda\mu s}(\theta, \varphi)), \quad (1)$$

where $c(\{\beta\})$ is the volume fixing factor, the real-valued spherical harmonics $Y_{\lambda\mu c}$, with $\mu > 0$ even and $Y_{\lambda\mu s}$,

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with $\mu > 0$ odd, are defined in terms of the usual ones as: $Y_{\lambda\mu c} = (Y_{\lambda\mu} + Y_{\lambda-\mu})/\sqrt{2}$ and $Y_{\lambda\mu s} = -i(Y_{\lambda\mu} - Y_{\lambda-\mu})/\sqrt{2}$. Such shapes have at least one symmetry plane $y-z$. Although nonaxiality modifies energy landscapes in the region of the third barrier, the main effect comes from a proper treatment of the axially symmetric shapes. Therefore, we explore here the first part of the Eq. (1).

Two models were used for the macroscopic part: the Yukawa plus exponential [25], used by us up to now in many studies, and recently given version of the Liquid Drop [26] with the mean-curvature term included in the deformation-dependent part of energy. The reason for using the latter was to check whether double third minima in [15–17] are not related to some singularity of the used Yukawa-plus-exponential macroscopic energy.

The s.p. potential parameters used in the present work, as well as the way the shell- and pairing corrections are the same as in a number of our previous studies, e.g. in [18–20]. Let us emphasize that the used model very well reproduces first [18], and second barriers [19] and second minima [20] in actinide nuclei. *Removing shape limitation* A general difficulty in studying very deformed nuclear shapes lies in their effective parametrization. The Eq. (1), natural for small and moderate deformations, becomes problematic for this purpose. Rather large (more than six) number of deformations is necessary for a sufficient accuracy. Their sizes may be misleading, for example, there are sets $\{\beta_\lambda\}$ corresponding to *huge* elongations with *quite moderate* β_2 . Such is the case of two kinds of IIIrd minima in actinides shown in [15–17]. With similar $\beta_2 \approx 0.85$ they might be thought close in elongation. But they are not: they correspond to different quadrupole moments $Q \approx 120$ and 170 b. The energy mapping in many-dimensional space becomes a problem. A reduction of dimension via the minimization over some of the deformations often leads to an energy surface composed from disconnected patches, corresponding to multiple minima in the auxiliary (those minimized over) dimensions. If a smooth look of such a surface hides the jumps between minima, it suggests a false picture of the barrier.

It may be for these difficulties that an important shape limitation of Eq. (1) did not attract much attention. The dipole deformation β_1 is omitted there, as corresponding to a shift of the origin of coordinates which leaves energy (always calculated in the center of mass frame) invariant. However, this is true only for *weakly* deformed shapes. For large elongations, β_1 acquires a meaning of a real shape variable.

The parametrization (1) is not unique, as the same shape has many expansions corresponding to various shifts of the origin of coordinates. Hence, one could think that β_1 may be always transformed out. However, the shift invariance is lost because a) the expansion (1) is always truncated, b) for well constricted shapes, large shifts lead outside the formula (1) making the radius mul-

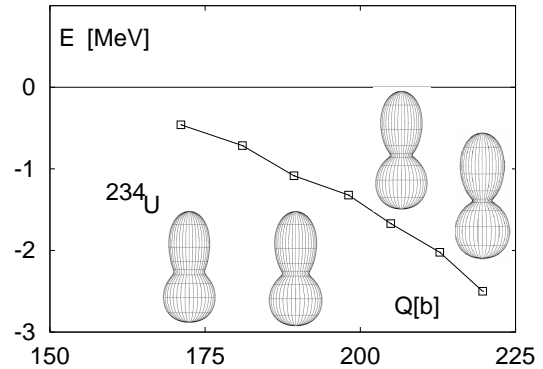


FIG. 1: Energy along a sequence of smoothly elongating shapes parametrized by $\beta_1 - \beta_8$, beginning at the apparent third minimum with larger octupole deformation $\beta_3 \approx 0.6$ in ^{234}U . Quadrupole moments calculated for the sharp-surface distribution and the radius parameter $r_0 = 1.16$ fm. Depicted shapes correspond to: $Q = 171$ (the apparent IIIrd minimum), 189, 205 and 220 b.

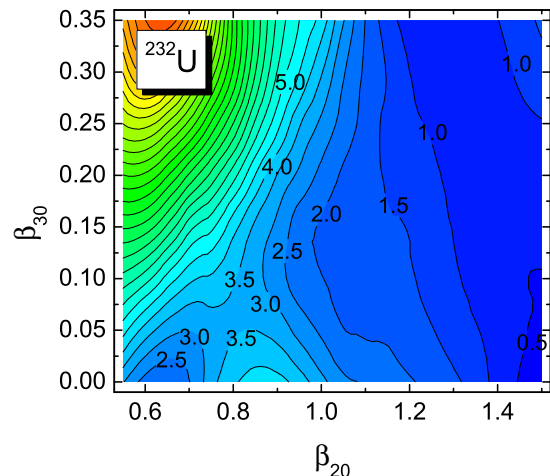
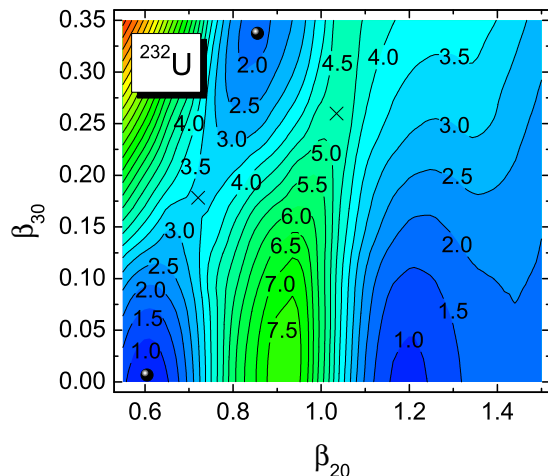
tivalued. In practice, admitting nonvanishing β_1 in (1),

$$R(\theta, \varphi) = c(\{\beta\})R_0(1 + \sum_{\lambda=1} \beta_\lambda Y_{\lambda 0}(\theta)), \quad (2)$$

turns out to be an effective way of including an important class of smoothly constricted elongated shapes, which are *unreachable* when using a reasonably truncated expansion (1) *without* it. One should notice that the parametrization (2) is redundant: the remaining invariance to small shifts of the origin of coordinates means that some shapes have many parametrizations with different β_1 .

Disappearance of spurious minima We considered even-even nuclei $^{230,232}\text{Th}$, $^{232,234,236,238}\text{U}$ and ^{240}Pu . Axially symmetric shapes were assumed. Two sets of energy calculations were done: A) maps (β_2, β_3) with minimization over $\beta_4-\beta_8$ (7D) and B) in a hypercube $\beta_1-\beta_6$ (6D) from which various 2D maps could be generated. The grid points were (with step in all directions = 0.05): $-0.35-0$ in β_1 , $0.55-1.5$ in β_2 , $0-0.35$ in β_3 , $-0.10-0.35$ in β_4 , $-0.2-0.2$ in β_5 , $-0.15-0.15$ in β_6 . To improve limits on barrier heights we calculated energies along chosen continuous paths in 8D space $\beta_1-\beta_8$.

Results of A) essentially reproduce landscapes in [15, 16]. Beyond the axially- and reflection-symmetric second minima, the reflection-asymmetry lowers the second saddles. Even more (shell correction of ≈ 10 MeV) it lowers energy for still larger elongations. Double IIIrd minima with $\beta_3 \approx 0.3$ and 0.6 are obtained with the Yukawa-plus-exponential macroscopic energy; the less mass-asymmetric ones are deeper in Th and higher in U and Pu. With the LSD macroscopic energy, formula [26] minima with larger β_3 (quadrupole moments) vanish



and the IIIrd minima become single. In both versions the third barriers are quite high, ≥ 2.5 MeV above the IIIrd minimum, with the highest ones in thorium.

The landscape modification obtained by including dipole deformation β_1 in (1) is decisive for the picture of the IIIrd minima. The apparent minima with large β_3 and quadrupole moments disappear completely in the studied nuclei. One can find continuous 8D paths starting at the supposed IIIrd minimum and leading to scission, along which energy decreases gradually. An example is shown in Fig. 1 for ^{234}U .

The existence of minima with smaller octupole deformation (and quadrupole moments) requires a more detailed study. Finding the barrier in many-dimensional space requires hypercube calculations. The maps (β_2, β_3) generated by minimization over β_4 - β_6 from the 6D hypercube calculations B) are shown in Fig. 2 for ^{232}U at fixed $\beta_1 = 0$ and -0.2 . The large effect of included new shapes may be seen as a lowering of the $\beta_1 = 0$ third barrier by $\beta_1 = -0.2$. Its size of 2-4 MeV is common in all studied nuclei, leaving some small barriers in $^{230,232}\text{Th}$. One can note that the heights of the second saddles are practically *not changed* by β_1 . Hence, in the evaluation of secondary fission barriers in actinides (see e.g [19]) this parameter is not necessarily needed. The results B) indicate a large effect of new shapes, but are not precise enough for a detailed barrier prediction. Our tests show that deformations β_1 - β_8 are needed for fixing minima up to 100-150 keV, a greater precision requires at least β_9 . As 8D or 9D hypercube calculations are very time-consuming, we find the upper limit on barriers selecting continuous paths in 8D β_1 - β_8 space, from the supposed

IIIrd minima towards scission. Energies vs quadrupole moments along such paths are shown in Fig. 3 for ^{232}Th and ^{232}U . As can be seen, the barrier vanishes in uranium and must be smaller than 330 keV in ^{232}Th . The only other nonzero upper limit on the IIIrd barrier of 200 keV we find in ^{230}Th . As by increasing the number of deformations one can only lower saddle points, the third barriers could rise only by lowering the supposed IIIrd minima. But from our tests this effect is of the order of 100 keV, so, within the used model, the barrier in ^{232}Th should not be higher than ≈ 450 keV. The results of this work may be summarized as follows:

(i) With the shape parametrization (1), modifications of energy landscapes by including the dipole deformation β_1 become important beyond the IInd barrier in actinides.

(ii) They remove minima with larger octupole deformations found in [15, 16]. They also remove minima with smaller octupole deformations, perhaps, except $^{230,232}\text{Th}$, where only a very shallow minima can exist. Time-consuming, 9D calculations are required for a precise (up to less than 100 keV) determination of the IIIrd barriers.

(iii) The IIIrd barriers predicted by the Woods-Saxon model, if they exist at all, do not exceed a few hundreds keV which is in agreement with selfconsistent mean-field results with realistic forces. They do not correspond to a type suggested originally in [15, 16] which served a physical interpretation of some experimental data.

It remains to understand the observed resonances in fission probability and their structure. On the theory side, with the mean-field barriers so shallow, one should

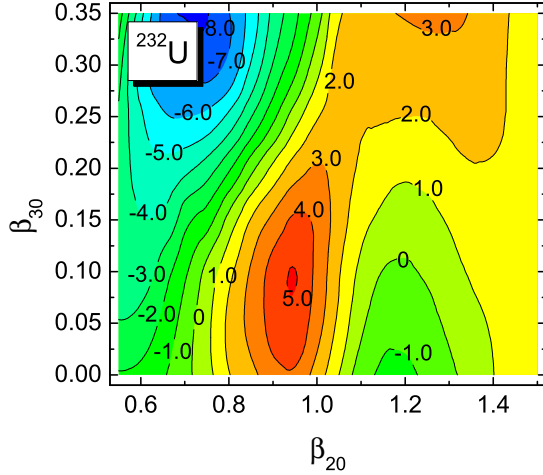


FIG. 2: Energy maps for ^{232}U from the 6D $\beta_1 - \beta_6$ calculation, minimized over β_4, β_5 ; upper panel - $\beta_1 = 0$, middle panel - $\beta_1 = -0.2$. The lower panel gives the difference $E_{\text{upper}} - E_{\text{middle}}$, so the lowering of the third barrier by $\beta_1 = -0.2$ can be read from it as a positive number.

check collective (beyond mean-field) corrections. One could also check barriers at higher spins.

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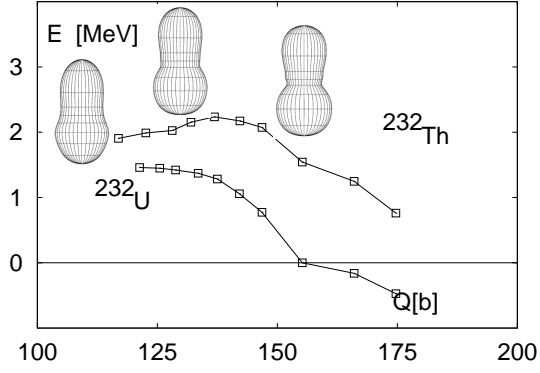


FIG. 3: Energy along a sequence of continuously elongating shapes parametrized by $\beta_1 - \beta_8$, beginning at the apparent third minima with smaller octupole deformation $\beta_3 \approx 0.3$ in ^{232}Th and ^{234}U . Quadrupole moments calculated as in Fig.1. Depicted shapes correspond to states in ^{232}Th : $Q=117$ (apparent IIIrd minimum), 137 and 155 b.